Manipulation of Cold Atomic Collisions by Cavity QED Effects.

J.I. Kim, R.B.B. Santos, and P. Nussenzveig Instituto de Física, Universidade de São Paulo, Caixa Postal 66318, CEP 05315-970, São Paulo, SP, Brazil. (Received 10 May 2000)

We show how the dynamics of collisions between cold atoms can be manipulated by a modification of spontaneous emission times. This is achieved by placing the atomic sample in a resonant optical cavity. Spontaneous emission is enhanced by a combination of multiparticle entanglement together with a higher density of modes of the modified vacuum field, in a situation akin to superradiance. A specific situation is considered and we show that this effect can be experimentally observed as a large suppression in trap-loss rates.

PACS numbers: 32.80.Qk, 34.20.Cf, 33.80.-b, 42.50.Fx

Experiments with cold and ultracold atoms have led to many recent achievements, such as Bose-Einstein condensation [1], atom optics and interferometry [2], and precision measurements [3]. Atomic collisions at these very low temperatures [4] are of great importance in many of these applications. The density of atoms attainable in optical traps is usually limited by exoergic inelastic collisions, which lead to trap loss. The study of these processes presents several interesting features, since the dynamics is very distinct from collisions at higher temperatures. Because the atoms move so slowly, not only are they sensitive to long-range interaction potentials but, in the presence of light, they can also undergo changes of internal states during a collision (the interaction time can be larger than the typical spontaneous emission time). Therefore, these collisions can be manipulated with light, as demonstrated, for example, in experiments of optical shielding [5] of trapped atoms from collisional loss.

One parameter, however, has been overlooked for the optical manipulation of cold collisions. The final outcome of a two-body encounter depends strongly on the spontaneous emission time. In this Letter we show, for the first time, how to modify spontaneous emission times in the context of cold collisions, and thereby manipulate the collisional dynamics. This is done by a combination of multiparticle entanglement [6] together with a modified vacuum field in Cavity QED [7], in a situation akin to superradiance [8–10]. We focus here on one specific collision process and analyze it in the presence of an optical cavity. Orders of magnitude of trap-loss probabilities show that this modification in collisional dynamics is within reach of current experimental techniques.

Let us describe briefly one of the first identified collisional loss processes, so-called radiative escape from a trap [4]. One atom of a colliding pair is excited by a laser of frequency ω_L at a large internuclear separation R, and the atoms are accelerated towards each other by the strong long-range dipole-dipole attractive potential $U = -C_3/R^3$, where C_3 is a constant that depends on the atom under consideration. If the spontaneous emis-

sion time is long, the pair may gain enough kinetic energy to escape from the trap (by emitting a photon with energy $\hbar\omega_{\gamma}$ smaller than that of the absorbed photon, $\hbar\omega_{L}$). The interaction potentials are sketched in Fig. 1 for the ground state nS and the excited state nP of an alkali atom. Two regions can be defined: $R < R_{e}$ and $R_{e} < R < R_{C}$, where R_{C} (the so-called Condon point, chosen by tuning ω_{L}) is the internuclear separation at which a weakly bound molecule is excited. The separation R_{e} is the smallest one for which spontaneous emission does not lead to trap loss. If one can enhance spontaneous emission of this excited molecule, decay will happen earlier, in the region between R_{C} and R_{e} in Fig. 1, preventing atoms from being lost.

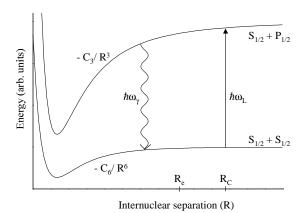


FIG. 1. Excited state long range dipole-dipole potential $U=-C_3/R^3$ and the ground state van der Waals attractive potential $-C_6/R^6$, C_6 being a constant. Its distance to the asymptote of U is the atomic separation $\hbar\omega_A$ between $nS_{1/2}$ and $nP_{1/2}$.

The modification of atomic radiative properties was one of the first effects to be demonstrated in Cavity QED [7]. Spontaneous emission enhancement [11,12] and inhibition [13] were demonstrated in the 80's. Radiative level shifts, such as a cavity-induced Lamb shift, were also demonstrated in this context [12,14]. However, spontaneous emission for single atoms in the optical domain was

not significantly enhanced, owing to the relatively small solid angle encompassed by a centimeter-sized Fabry-Perot cavity. It is, nevertheless, possible to achieve a large enhancement of spontaneous emission when we consider a sample of many identical weakly bound molecules (so-called *quasimolecules* [4]) coupled to the same cavity mode. The different excited quasimolecules are indistinguishable when interacting with the cavity field. Quantum interference will thence be important in the process of spontaneous emission into the cavity, in a way analogous to superradiance [8–10]. Since, for most experiments in cold collisions, the typical separation between different quasimolecules is greater than an optical wavelength, this interference will only be constructive if the quasimolecules are excited into a multiparticle entangled state by a laser beam injected into the cavity mode [9]. Cavity QED effects on cold atoms have been recently investigated for high-Q cavities interacting with single pairs of atoms in the context of cold collisions [15], and with large numbers of atoms in the context of forces exerted on the atoms [16].

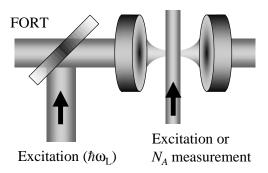


FIG. 2. Sketch of a possible experiment to probe Cavity QED-modified cold collisions. The N_A atoms are held near the center of a Fabry-Perot cavity by a FORT laser. The excitation (probe) laser, of frequency ω_L , can be sent either perpendicular to the cavity axis or via the cavity mode.

We consider a sample of cold atoms trapped near the center of a quasi-confocal optical resonator. In order to avoid optical pumping effects, atoms can be trapped in the ground state. This can be done, for instance, if the atoms are in a far-off resonance trap (FORT) [17], as sketched in Fig. 2, or in a magnetic trap [18]. The experiment would require highly reflective mirror coatings with a sharp edge, in order to transmit the FORT beam (detuned by a few nanometers from the atomic transition). However, the whole setup would be far from trivial, since it is not straightforward to load a FORT (from a Magneto Optical Trap, MOT [19]) inside an optical cavity in view of alignment difficulties. The dipole-dipole potential Uis turned on by an excitation laser red detuned from the atomic transition (separation $\hbar\omega_A$ between states $nS_{1/2}$ and $nP_{1/2}$). Our orders of magnitude will be calculated

for ^{85}Rb atoms, so the atomic ground and excited states in the following will be $5S_{1/2}$ and $5P_{1/2}$, respectively.

A colliding pair subject to a weak excitation field is likely to undergo just one-photon processes. The pairs are treated in the two-level approximation (which is justified later in the text) with a ground state $|g\rangle$ and an excited state $|e\rangle$ connected to the $5S_{1/2} + 5S_{1/2}$ and $5S_{1/2} + 5P_{1/2}$ asymptotic states, respectively. Here, the pair in state $|g\rangle$ is not bound but, in state $|e\rangle$, there is a weak binding force. For simplicity, we will refer to these pairs as quasimolecules, independent of their state $(|e\rangle)$ or $|g\rangle$). For a given R, the energy separation between $|g\rangle$ and $|e\rangle$ is $\hbar\omega_R = \hbar\omega_A - C_3/R^3$ (Fig. 1). Each quasimolecule interacts with the electromagnetic field with a dipole moment $(\sigma_i + \sigma_i^{\dagger}) \mathbf{d}_i$, where σ_i and σ_i^{\dagger} are Pauli operators acting in the subspace spanned by $|e\rangle$ and $|g\rangle$. The dipole \mathbf{d}_i , determined by the molecular axis [20], is randomly oriented with respect to the cavity field polarization. Its magnitude is $|\mathbf{d}_i| = \sqrt{2} d_A$, where d_A is the atomic dipole moment, with a resulting decay constant Γ which is twice that of the atomic excited state Γ_A [4].

When the quasimolecules are excited via the cavity mode, they end up in a multiparticle entangled state. We will only treat here the simplest entangled state, produced when a *single* excitation is injected into the cavity. In the ground state $|G; 1_{\mathbf{k}_L}\rangle \equiv |gg \cdots g; 1_{\mathbf{k}_L}\rangle$ all quasimolecules are in state $|g\rangle$ and there is one laser photon in the cavity. The quasimolecule-field interaction couples this state to all singly-excited states $|i;0\rangle$ ($|1;0\rangle \equiv$ $|eg\cdots g;0\rangle, |2;0\rangle \equiv |ge\cdots g;0\rangle$, and so on). Using the dipole and rotating-wave approximations [7], the matrix elements of the interaction hamiltonian H_{int} are given by $V_i = \langle i; 0 | H_{\rm int} | G; 1_{\mathbf{k}_L} \rangle = \mathcal{E}(\omega_L) f^c(\mathbf{r}_i) \boldsymbol{\epsilon}_L \cdot \mathbf{d}_i$. They depend on the positions \mathbf{r}_i of the quasimolecules along the cavity mode, characterized by a profile $f^{c}(\mathbf{r})$, polarization ϵ_L and field per photon $\mathcal{E}(\omega) = (2\pi\hbar\omega/\mathcal{V})^{1/2}$, where $\mathcal V$ is the effective mode volume. The entangled state produced is finally given by $|E;0\rangle = \sum_{i} V_{i}|i;0\rangle/\hbar\tilde{\Omega}$, defining the collective Rabi frequency $\tilde{\Omega} = (\sum_i |V_i|^2)^{1/2}/\hbar$. Note that although a single excitation is present, it is shared by all pairs.

We now calculate the spontaneous emission rate Γ_c for this entangled state. Summing over the final states $|G; 1_{\mathbf{k}}\rangle$ and considering all possible wavevectors \mathbf{k} for the emitted photon,

$$\frac{\Gamma_c}{2\pi} = \int d\omega_{\mathbf{k}} d\Omega_{\mathbf{k}} \ \rho \ \delta(\omega_{\mathbf{k}} - \omega_R) \frac{\sum_{\lambda} |\mathbf{X}_{\mathbf{k}} \cdot \boldsymbol{\epsilon}_{\lambda}|^2}{\hbar^2} \ , \quad (1)$$

where $\mathbf{X}_{\mathbf{k}} \equiv \left(\sum_{i} \mathcal{E}(\omega_{\mathbf{k}}) f_{\mathbf{k}}(\mathbf{r}_{i}) V_{i}^{*} \mathbf{d}_{i} / \hbar \tilde{\Omega}\right)$ and $f_{\mathbf{k}}(\mathbf{r})$ is the mode function for a given \mathbf{k} . The product $|\mathbf{X}_{\mathbf{k}} \cdot \boldsymbol{\epsilon}_{\lambda}|$ is the absolute value of the collective coupling between the quasimolecules and the field (matrix element of H_{int}). Taking into account that only emission into the solid angle $\Delta \Omega_{c}$ encompassed by the cavity mirrors is affected by the enhanced spectral density $\rho(\omega) = \rho_{0}(\omega)\Lambda(\omega)$ (for the

degenerate longitudinal modes), we separate its contribution from emission into the rest of free space. Here, $\rho_0(\omega)$ is the free space density and $\Lambda(\omega)$ is the cavity line shape function [21]. In view of the large number of degenerate transverse modes in the cavity, the integral over $d\Omega_{\bf k}$ can be replaced by a summation over transverse TEM_{nm} modes with profiles $f_{nm}({\bf r})$ [21]. The solid-angles $\Delta\Omega_{nm}$ encompassed by them are determined by their transverse dimensions at the mirrors. Diffraction losses are also accounted for, by substituting effective Λ_{nm} values for each TEM_{nm} mode. This correction is significant only for high-order modes, for which $\Delta\Omega_{nm} \sim \Delta\Omega_c$. An effective solid angle can be defined by

$$\Delta\Omega_{eff} \equiv \sum_{nm} \Delta\Omega_{nm} \frac{\Lambda_{nm}}{\Lambda_{00}} \frac{|\frac{1}{N} \sum_{i} |\boldsymbol{\epsilon}_{L} \cdot \mathbf{d}_{i}|^{2} f_{nm}^{*}(\mathbf{r}_{i}) f^{c}(\mathbf{r}_{i})|^{2}}{\frac{1}{N} \sum_{i} |\boldsymbol{\epsilon}_{L} \cdot \mathbf{d}_{i}|^{2} |f^{c}(\mathbf{r}_{i})|^{2}},$$
(2)

where N is the number of entangled quasimolecules. For typical optical cavities, this solid angle will be relatively small. Emission into the rest of free space will then be little affected by the presence of the cavity and it can be calculated to be approximately Γ [22]. For resonant excitation $\omega_R \sim \omega_L$ ($|\omega_R - \omega_L| \lesssim \gamma_c/2$, where $\gamma_c/2\pi$ is the cavity linewidth), and using $\Gamma = 2\Gamma_A = 8d_A^2\omega_A^3/3\hbar c^3$, we obtain

$$\Gamma_c \approx \left(1 + \frac{3}{2} \frac{\Delta \Omega_{eff}}{4\pi} N \Lambda_{00} \frac{\omega_L^3}{\omega_A^3}\right) \Gamma.$$
 (3)

To verify whether this can be a significant enhancement of spontaneous emission, we introduce realistic experimental parameters. We consider a (quasi-)confocal cavity with mirrors of diameter 2b = 1.0 cm and reflectivities r = 0.97 separated by $\ell = 2.9$ cm. The excitation field, with circular polarization, matches a TEM_{00} mode. The cavity linewidth is then $\gamma_c/2\pi \approx 200$ MHz and the line shape function is $\Lambda_{00} = 66$, with a corresponding finesse $\mathcal{F} = 103$. We still need to determine the number N of entangled quasimolecules and consider their distribution along the cavity mode. We assume that our sample of cold ⁸⁵Rb atoms ($\lambda_A = 2\pi c/\omega_A = 795$ nm and $\Gamma_A/2\pi = 6$ MHz) is trapped in a FORT (see Fig. 2), near the center of the cavity, in a cigar-shaped cloud with length $L \approx 0.6$ mm and radius $a \approx 2.6 \times 10^{-2}$ mm. The number N of quasimolecules in the state $|E;0\rangle$, for a detuning $\delta = \omega_L - \omega_A = -2\pi \times 100$ MHz, can then be estimated by counting all pairs such that $U(R_C) - U(R) \le$ $\hbar\Gamma$, since the excitation laser linewidth ($\sim 1 \text{ MHz}$) is negligible. With $C_3 = 11.4 \times 10^{-11} \text{ erg Å}^3$, this gives a spread $\Delta R \sim \hbar \Gamma / |U'(R_C)| \sim 22.4 \text{ Å about } R_C \simeq 556 \text{ Å}.$ For $N_A \sim 10^6$ atoms at a density $n_A \sim 10^{12} \ {\rm cm}^{-3}$ we have $N \simeq \frac{1}{2} N_A n_A 4\pi R_C^2 \Delta R \sim 45$ pairs. Even though this is a relatively crude approximation, attaining a number of this order should be feasible, since even larger numbers N_A of atoms in a FORT were recently reported by

Corwin et al. [23]. Moreover, if the experiment were to be performed in a magnetic trap, N_A could be larger by a few orders of magnitude. The distribution of quasimolecules in the cavity is simulated by sorting out 10 sets of 45 random positions \mathbf{r}_i and dipole orientations \mathbf{d}_i . After averaging, we obtain $\Delta\Omega_{eff}/4\pi=7.4\times10^{-4}$ and $\Gamma_c/\Gamma\approx4.3$.

Such an enhancement of spontaneous emission would lead to observable consequences, as in the process of radiative escape of atoms from the trap. This process does not occur naturally in a FORT, since the atoms are trapped in the ground state. It has to be induced by a separate probe laser, as in the photoassociation experiments by Cline et al. [24]. If this laser beam is sent via the cavity mode, an entangled state will be excited and the spontaneous emission rate will show collective enhancement. On the other hand, if it propagates perpendicular to the cavity axis (see Fig. 2), the interactions with individual quasimolecules will not be indistinguishable and the spontaneous emission rate will be at most cavity-enhanced, thus close to Γ [12]. We can therefore compare directly these two situations and measure trap losses "with" and "without" cavity.

The trap-loss probability will be calculated using semiclassical models [4,25,26], for which the probability of excitation is treated independently from emission. These models have been shown to give good results in the regime of low excitation laser intensities (we consider only a single excitation in the cavity) and for detunings $\delta \gtrsim 10\Gamma_A$ [26]. We describe the specific transition chosen here $(5S_{1/2} + 5S_{1/2} \rightarrow 5S_{1/2} + 5P_{1/2})$ asymptotic states) as a two-level system. As shown by Peters et al. [27], multilevel crossings are not important to this transition for our detuning range. The net result is an average over several similar hyperfine-splitted two-level systems, which would not lead to substantial modifications in the orders of magnitude we calculate. All these models deal only with the trap-loss probability of single quasimolecules but, as we show below, they can be adapted to consider an excited entangled state.

In order to estimate trap losses, we consider a trap depth V_0 of the FORT of the order of 5 mK (~ 100 MHz). The kinetic energy gained has then to be greater than 10 mK for both atoms of the quasimolecule to escape from the trap. This places an upper bound for the cavity linewidth $\gamma_c/2\pi \leq 2V_0 \sim 200$ MHz, that defines the resonant region $R_e < R < R_C$ where emission does not lead to trap loss. As the atoms of a colliding pair accelerate towards each other, the decay rate of $|E;0\rangle$ shifts from Γ_c to the off-resonance value Γ [22], since R for each component state $|i;0\rangle$ of $|E;0\rangle$ decreases below R_e . After this first passage in the region $R < R_e$, the probability that two atoms, from any quasimolecule i, escape from the trap is $\sum_{i} |V_i/\hbar \tilde{\Omega}|^2 (1 - e^{-2t_e \Gamma}) e^{-t_c \Gamma_c}$, where t_c (t_e) is the time interval spent between R_C and R_e (R_e and R=0). Notice that $\sum_{i} |V_i/\hbar \tilde{\Omega}|^2 = \langle E; 0|E; 0 \rangle = 1$, so

the difference from the expression obtained "without cavity" is the existence of two radiative damping rates Γ and Γ_c . The vibrational levels of U(R) are accounted for by allowing multiple-passages across R_e [27] before emission occurs (for our detuning, these levels are not resolved and a wavepacket containing several levels is excited [25–27]). The overall loss-probability \mathcal{L}_c is then

$$\mathcal{L}_c = \sum_{i=1}^N \left| \frac{V_i}{\hbar \tilde{\Omega}} \right|^2 \sum_{n=1}^\infty (1 - e^{-2\Gamma t_e}) e^{-(2n-1)\Gamma_c t_c - 2(n-1)\Gamma t_e}$$

$$= \frac{\sinh(t_0 - t_c)\Gamma}{\sinh[t_0 + (\Gamma_c/\Gamma - 1)t_c]\Gamma} \equiv p \mathcal{L}_0, \tag{4}$$

where $\mathcal{L}_0 \equiv \sinh{(t_0 - t_c)} \Gamma / \sinh{t_0} \Gamma$ is the cavity-free loss-probability and $t_0 = t_c + t_e = 3.0 \times 10^{-8} \,\mathrm{s} \,(\sim \Gamma_A^{-1})$ is the total time interval from R_C to R = 0, obtained from conservation of energy $\mu \dot{R}^2 / 2 + U(R) = \mathrm{const} \, [25]$, neglecting the initial velocity \dot{R} at R_C . Here μ is the reduced mass of the colliding pair. Approximating $\sinh(x) \approx e^x / 2$ for $x \geq t_0 \Gamma \approx 2.3$, we see that $\mathcal{L}_c / \mathcal{L}_0 = p \approx e^{-(\Gamma_c - \Gamma)t_c}$ is simply limited by the ratio between the survival probabilities, in the first passage through the region $R_e < R < R_C$, with and without cavity. In order to measure $\mathcal{L}_c / \mathcal{L}_0$, we can adjust the laser intensities to have the same fraction of excited-state quasimolecules in both situations (excitation via cavity mode or excitation beam perpendicular to the cavity axis). The excitation probability factors out, and we obtain with our parameters $(\Gamma t_c \approx 1.7) \, \mathcal{L}_c / \mathcal{L}_0 \approx 3 \times 10^{-3}$.

This large predicted trap loss suppression should encourage future attempts to observe it experimentally. Other experimental setups could have been considered, in which larger trap depths and numbers of atoms are obtained (such as magnetic traps), leading to even more encouraging orders of magnitude. Our results could also be extended to a situation with a larger fraction of excited molecules, closer to the superradiant regime [8–10]. One must notice, however, that this is not a method directly applicable to suppressing trap loss in a MOT, for example. Quasimolecules must be excited via the cavity mode, which would not happen in most existing optical traps. On the other hand, trap loss may conceivably be prevented in novel traps using optical resonators, as proposed in [28].

In summary, we have shown, for the first time, that collisions between cold atoms can be manipulated by controlling the spontaneous emission time. This is achieved through multiparticle entanglement in a cavity-modified electromagnetic vacuum. Orders of magnitude of traploss probabilities show that this effect may be experimentally observed with present-day technology.

The authors acknowledge helpful discussions with K.L. Corwin, D. Kleppner, A. Lezama, R. Napolitano, H.M. Nussenzveig and M. Raizen, and financial support from FAPESP and CNPq.

- M.H. Anderson et al., Science 269, 198 (1995); K.B. Davies et al., Phys. Rev. Lett. 75, 3969 (1995); C.C. Bradley et al., Phys. Rev. Lett. 75, 1687 (1995).
- [2] C.S. Adams, O. Carnal, and J. Mlynek, Adv. At. Mol. Opt. Phys. 34, 1 (1994).
- [3] See, e.g., C. Fertig and K. Gibble, Phys. Rev. Lett. 85, 1622 (2000).
- [4] See, e.g., J. Weiner et al., Rev. Mod. Phys. 71, 1 (1999).
- [5] L. Marcassa et al., Phys. Rev. Lett. 73, 1911 (1994); S. Bali et al., Europhys. Lett. 27 273 (1994); V. Sanchez-Villicana et al., Phys. Rev. Lett. 74, 4619 (1995).
- [6] C.A. Sackett et al., Nature (London) 404, 256 (2000);
 D. Bouwmeester et al., Phys. Rev. Lett. 82, 1345 (1999);
 A. Rauschenbeutel et al., Science 288, 2024 (2000); and references therein.
- [7] S. Haroche, in Les Houches, Session XXXVIII, 1982, edited by G. Grynberg and R. Stora, Vol.I (North-Holland, Amsterdam, 1984).
- [8] R.H. Dicke, Phys. Rev. 93, 99 (1954).
- [9] R. Bonifacio, P. Schwendimann, and F. Haake, Phys. Rev. A 4, 302 (1971); 4, 854 (1971).
- [10] M. Gross and S. Haroche, Phys. Rep. 93, 301 (1982).
- [11] P. Goy et al., Phys. Rev. Lett. 50, 1903 (1983).
- [12] D.J. Heinzen *et al.*, Phys. Rev. Lett. **58**, 1320 (1987);
 D.J. Heinzen and M.S. Feld, *ibid.* **59**, 2623 (1987).
- [13] H.G. Hulet, E.S. Hilfer, and D. Kleppner, Phys. Rev. Lett. 55, 2137 (1985).
- [14] M. Brune, P. Nussenzveig, F. Schmidt-Kaler, F. Bernardot, A. Maali, J.M. Raimond, and S. Haroche, Phys. Rev. Lett. 72, 3339 (1994).
- [15] B. Deb and G. Kurizki, Phys. Rev. Lett. 83, 714 (1999).
- [16] M. Gangl and H. Ritsch, Phys. Rev. A 61, 043405 (2000).
- [17] S. Chu et al., Phys. Rev. Lett. 57, 314 (1986); J.D. Miller,
 R.A. Cline, and D. J. Heinzen, Phys. Rev. A 47, R4567 (1993).
- [18] V. Bagnato et al., Phys. Rev. Lett. 58, 2194 (1987).
- [19] E.L. Raab et al., Phys. Rev. Lett. 59, 2631 (1987); C. Monroe et al., ibid. 65, 1571 (1990).
- [20] R. Napolitano, J. Weiner, and P.S. Julienne, Phys. Rev. A55, 1191 (1997).
- [21] A. Yariv, Quantum Electronics (John Wiley & Sons, New York, 1989) 3rd ed.; see also [12].
- [22] Details will be given in J.I. Kim, R.B.B. Santos, and P. Nussenzveig, in preparation.
- [23] K.L. Corwin et al., Phys. Rev. Lett. 83, 1311 (1999).
- [24] R.A. Cline, J.D. Miller, and D.J. Heinzen, Phys. Rev. Lett. 73, 632 (1994); J.D. Miller, R.A. Cline, and D.J. Heinzen, *ibid.* 71, 2204 (1993).
- [25] A. Gallagher and D.E. Pritchard, Phys. Rev. Lett. 63, 957 (1989).
- [26] K.-A. Suominen et al., Phys. Rev. A57, 3724 (1998).
- [27] M.G. Peters et al., Phys. Rev. A50, R906 (1994).
- [28] V. Vuletić and S. Chu, Phys. Rev. Lett. 84, 3787 (2000).